

Electrical conductivity of viscous liquid foods

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Abstract

The electrical conductivities of foods are used for quality assurance, electrical tomography studies and are required for effective simulation of electrical heating processes such as ohmic and microwave heating. Solutions containing milk solids, whey proteins, sugars and sodium carboxymethyl cellulose (NaCMC), with and without electrolytes were prepared and tested. The electrical conductivity was measured using an RCL meter connected to a parallel plate probe. At low concentrations the conductivity increased with concentration, but in some viscous solutions the reduced ion mobility caused a drop in conductivity. The conductivity of sugar solutions could be related to following the modified Walden equation, but that of NaCMC solutions was not influenced by the bulk viscosity. Instead an ion “diffusion viscosity” was defined and calculated from the conductivity. It was found to correspond to the likely viscosity of the solution at a molecular scale.

Keywords

milk; whey protein; sucrose; glucose; carboxymethyl cellulose; ion mobility

1 Introduction

Foods such as honey, milk, and yogurt are solutions and colloidal dispersions containing carbohydrates, fat, sugars, proteins, minerals, and other minor components in water. Knowledge of physical properties is required for accurate design and simulation of processes and in this study electrical conductivity is the main property of interest, especially in the way it is affected by viscosity.

Sharifi & Young (2012, 2013) measured the conductivity of milk solutions with up to 47% solids and used the results for electrical tomography. They used multiple linear regression to relate the solids content and temperature to electrical conductivity, but they did not consider the effect of viscosity directly. St-Gelais et al. (1995) measured conductivity and viscosity of milk solutions as the pH was reduced in an attempt to monitor gelation. While their data show that both conductivity and viscosity changed, the variables appeared to be independent of each other. When the pH changed rapidly from 5.6 to 5.0, the viscosity increased about 100 times, while the conductivity only doubled. Henningsson et al. (2005) stated that proteins and lactose affected electrical conductivity of milk via viscosity, and that the charged proteins contributed to only 0.5% of conductivity by carrying charge. They suggested that the effect of proteins on viscosity and hence on conductivity was important.

In liquid foods, with the exception of a few such as soy sauce and fish sauce, the concentration of electrolytes is relatively low. For example concentrated skim milk with 50% solids content will contain only about 5% salts and organic acids only some of which are present as dissociated ions (Walstra, Wouter & Geurts, 2006), and honey has at most 2% ash and free

acids (White et al. 1962). Tomato ketchup which contains cellulose and starches has a maximum of about 5% NaCl, KCl and acids (Sharoba et al., 2005).

The starting point for most analyses is Walden's (1906) rule that proposes that electrical conductivity of a solution is inversely proportional to its viscosity. A number of researchers have studied saccharide + electrolyte systems and have proposed modifications to Walden's rule. Stokes (1959) confirmed that for KCl + sucrose solutions conductivity was inversely proportional to viscosity raised to the power of 0.7. Miller et al. (2000) reported powers of 0.64 for NaCl in trehalose and 0.78 for glucose. Longinotti and Corti (2002) gave similar results for MgCl₂, CaCl₂, NaCl and KCl in trehalose and sucrose, which were obtained when studying the cryoprotection properties of these solutions. They suggested changes in conductivity with concentration could be related to "preferential solvation, which in turns developed structural inhomogeneities". More data is given for various chlorides in sucrose by Mitra et al. (2010).

Acquarone et al. (2007) used electrical conductivity with pH at different dilutions of honey to discriminate the geographical origin of honeys. Bordi and Cametti (1986) showed that the concentration (and hence the bulk viscosity) of carboxymethyl cellulose (NaCMC) at up to 1 g/L has little effect on electrical conductivity when the concentration of added BaCl₂ was greater than about 3 mmol/L. Below that concentration they used Manning ion condensation concepts to explain the effect of NaCMC concentration. The viscosities of the samples were likely to be up to 5 times that of water.

This study was conducted to establish relationship for a range of food liquids at higher concentrations and hence higher viscosities. Model solutions were prepared using sugars, whey protein, milk powder, carboxymethyl cellulose and salts, and the electrical conductivity and viscosity of these was measured.

2 Theory

At low concentration the conductivity, σ , of a solution can be written as a sum for different ionic species

$$\sigma = \sum_i C_i \lambda_i \quad (1)$$

where C_i is molar concentration of ion i , and λ_i is the ionic conductivity which in turn can be defined in terms of Faraday's number, F , ion charge, z_i , and ion mobility, u_i .

$$\lambda_i = F z_i u_i \quad (2)$$

Ion mobility can be related to diffusion and hence to viscosity, η using the Walden equation.

$$u_i = \frac{e_0 z_i}{6\pi\eta r_i} \quad (3)$$

Here e_0 is the charge of an electron and r_i is the Stokes' radius.

For saccharide + electrolyte systems it has been found that as the viscosity increases $\lambda \propto 1/\eta^\alpha$ where α is a constant that has been determined to be in the range 0.64 to 0.78 for various sugars. Hence the modified Walden equation is written.

$$u_i = \frac{e_0 z_i}{6\pi\eta^\alpha r_i} \quad (4)$$

But to be dimensionally correct, the viscosity term should be written as $\eta_0(\eta/\eta_0)^\alpha$ where η_0 is a reference viscosity in consistent units.

For binary aqueous electrolyte solutions, as the salt concentration, C , increases the calculated conductivity decreases and is often described by Kohlrausch's Law

$$\sigma = C\Lambda_m = C(\Lambda_m^0 - KC^{0.5}) \quad (5)$$

where Λ_m and Λ_m^0 are the molar conductivity of the solute at concentration C and at infinite dilution respectively, and K is a constant. Using this as a basis, and with data from Chambers, Stokes and Stokes (1956) at 25 °C for concentrations up to 5.4 mol L⁻¹ (26%), the molar conductivity of NaCl is given at 20 °C by

$$\Lambda_{M,NaCl} = (114.6 - 28.6C_{NaCl}^{0.5}) \frac{\eta_w(25)}{\eta_w(T)} \quad (6)$$

Here molar conductivity is in units of mS.L.cm⁻¹mol⁻¹ and concentration is in mol L⁻¹. For KCl up to 4 mol L⁻¹ the equation is:

$$\Lambda_{M,KCl} = (144.3 - 32.6C_{KCl}^{0.3}) \frac{\eta_w(25)}{\eta_w(T)} \quad (7)$$

These equations (5 – 7) do not separate the effect of viscosity changes as concentration increases. Analysis of the data of Chambers et al. (1956) shows that about 90% of the drop in molar conductivity is due to the increase in viscosity over the concentration range.

van Rysselberghe and Nutting (1934) showed that adding NaCl and KCl conductivities on a molar basis was satisfactory if the component (binary) conductivities, $\Lambda_{m,i}$, are calculated at the total salt concentration, ΣC_i .

$$\sigma_{mix} = \Lambda_{m,mix} \Sigma C_i = \Sigma (C_i \cdot \Lambda_{m,i}) \quad (8)$$

Combining various equations in a more general form for a vector of concentrations in a mixed solution, \mathbf{C} , we can explicitly separate the effect of viscosity from the effect of increased ion concentration using the form:

$$\sigma = \frac{f(\mathbf{C})}{\eta} \quad (9)$$

It is proposed to use data from dilute binary solutions with similar ion concentrations as in the liquid foods, but with low, known viscosities to obtain estimates of $f(C)$ for each of the components in a solution and combine these using an equation of the form of (8). In food systems the exact composition is often unknown so it is intended obtain an effective concentration of all salts lumped together as single equivalent salt such as NaCl.

It is known that the inverse relationship with viscosity in Equation (3) will fail when solutions are no longer dilute, but rather than use the modified Walden equation (4), it is proposed that an effective viscosity be determined. This will be the viscosity experienced by conducting ions as they diffuse under the influence of the electric field. In this work it is termed the “diffusion viscosity”, η_{diff} , which can be estimated from the known ion concentrations and the measured conductivity.

$$\eta_{diff} = \frac{f(\mathbf{C})}{\sigma} \quad (10)$$

The bulk viscosities, η , of binary solutions of glucose, fructose and sucrose were required to calculate conductivities using the modified Walden equation (4). Data were obtained from Weast (1977) and equations were fitted in the form of Equation (11) recommended by Morison and Hartel (2007) to obtain constants.

$$\eta(T) = \eta_w(T) e^{\sum_i a_i \frac{w_i}{w_w} + b_i \left(\frac{w_i}{w_w}\right)^2} \quad (11)$$

Here T is the temperature, w is mass fraction, a and b are temperature dependent constants for a binary solution, subscript i refers to a single component i and subscript w refers to water. Table 1 gives a set of coefficients for this equation.

Table 1. Temperature ($^{\circ}\text{C}$) dependent coefficients for viscosity using Equation (11)

Component	a	b
Sucrose	$3.165 - 0.01956T + 6.9 \times 10^{-5}T^2$	$0.0063 - 0.0037T + 2.2 \times 10^{-5}T^2$
Glucose	$2.829 - 0.0126T - 1.2 \times 10^{-4}T^2$	$0.0097 - 0.0045T$
Fructose	$2.764 - 0.0177T + 9.4 \times 10^{-5}T^2$	$0.082 - 0.0064T$

3 Materials and Methods

Skimmed milk solids purchased from a local supermarket (Alpine brand, 55% lactose, 33% protein, 1% fat, 7% minerals, 4% water) was used to prepare milk solutions. A stick homogenizer was used to disperse the milk solids in Milli-Q water in the preparation of milk solutions. The solutions were held at 45°C for 40 minutes to enable hydration. Binary solutions of glucose, fructose and sucrose were prepared from food grade sugars. Sodium carboxymethyl cellulose (NaCMC) (Walocel C, DOW) solutions were prepared at concentrations up to 1% by mass of NaCMC powder (wet basis). The powder was dried at 105°C and found to contain 8.5% water. In addition various concentrations of KCl, NaCl and sucrose were added. Solutions of whey protein isolate (WPI) (Balance Sports Nutrition, New Zealand), with and without lactose and NaCl were also prepared. Weighing was achieved with an accuracy better than 1 mg in 100 g of solution. The set of solutions tested is given in Table 2. The preparation and conductivity of NaCMC solutions was repeated two months after the first set.

Table 2. Composition of solutions

Main component	Mass fraction	Added component	Mass fraction
NaCMC	0 – 1%		
NaCMC	0 – 1%	NaCl	0.01%
NaCMC	0 – 1%	KCl	0.01%
NaCMC	0 – 1%	Sucrose	30%
		KCl	0.013%
WPI	1, 10, 20, 30%		
WPI	20%	NaCl	0.01%, 0.1%, 1%
WPI	20%	Lactose	20%
		NaCl	0, 0.01%, 1%
Fructose	0.01% - 55%		
Glucose	0.01% - 60%		
Sucrose	0.1% - 70%		
Skim milk reconstituted	1% - 45%		

The electrical conductivity of the solutions were measured using a Schott conductivity probe LF413T, with an electrode gap of about 1 cm, connected to a Schott Lab 960 benchtop meter, the same probe was connected to a FLUKE PM6306 programmable RCL meter operating at

0.1 V a.c.. The RCL meter was used because the benchtop meter had a minimum resolution of 0.1 $\mu\text{S}/\text{cm}$ which was insufficient at the low conductivities measured in concentrated sugar solutions. A custom-made sample holder was used to seal the conductivity probe in place and to provide consistent experimental conditions. The container was immersed in a water bath at 20.0 $^{\circ}\text{C}$ with control of ± 0.02 $^{\circ}\text{C}$. The temperature was measured with a precision platinum thermometers with an accuracy of ± 0.01 $^{\circ}\text{C}$. The resistance at a frequency with zero or near zero phase angle was selected for the conductivity calculations. This reduced the influence of any capacitance or inductance in the measurement system. For low conductivity solutions the best frequency was about 100 Hz, but it was as high as 70 kHz for highly conductive solutions. The error introduced by doubling or halving the frequency was less than 2% in all cases, indicating low sensitivity to frequency. The cell was calibrated using standard KCl solutions with different mass fractions (Shreiner and Pratt, 2004) with a maximum deviation of 1%. Repeated measurements were found to be always consistent to better than the larger of 0.5% or 1.5 $\mu\text{S}/\text{cm}$. When the temperature deviated from 20.0 $^{\circ}\text{C}$, by at most 0.02 $^{\circ}\text{C}$, the measured resistance were corrected to 20.0 $^{\circ}\text{C}$ using a measured temperature coefficient of 2% $^{\circ}\text{C}^{-1}$. Repeated calibration of the conductivity and viscosity measurements showed no measureable drift.

For all solutions, a Haake Rotovisco RV20 with an NV cylinder set was used to measure the viscosities at 20.0 $^{\circ}\text{C}$. The Haake viscometer was calibrated using a Cannon N1000 calibration oil to within $\pm 1\%$ over the rotational speed range of the viscometer. At absolute uncertainty was estimated from water to be better than 0.3 mPa.s. Repeated measurements were always less than 2% different. The apparent viscosities are given at a shear rate of 2700 s^{-1} .

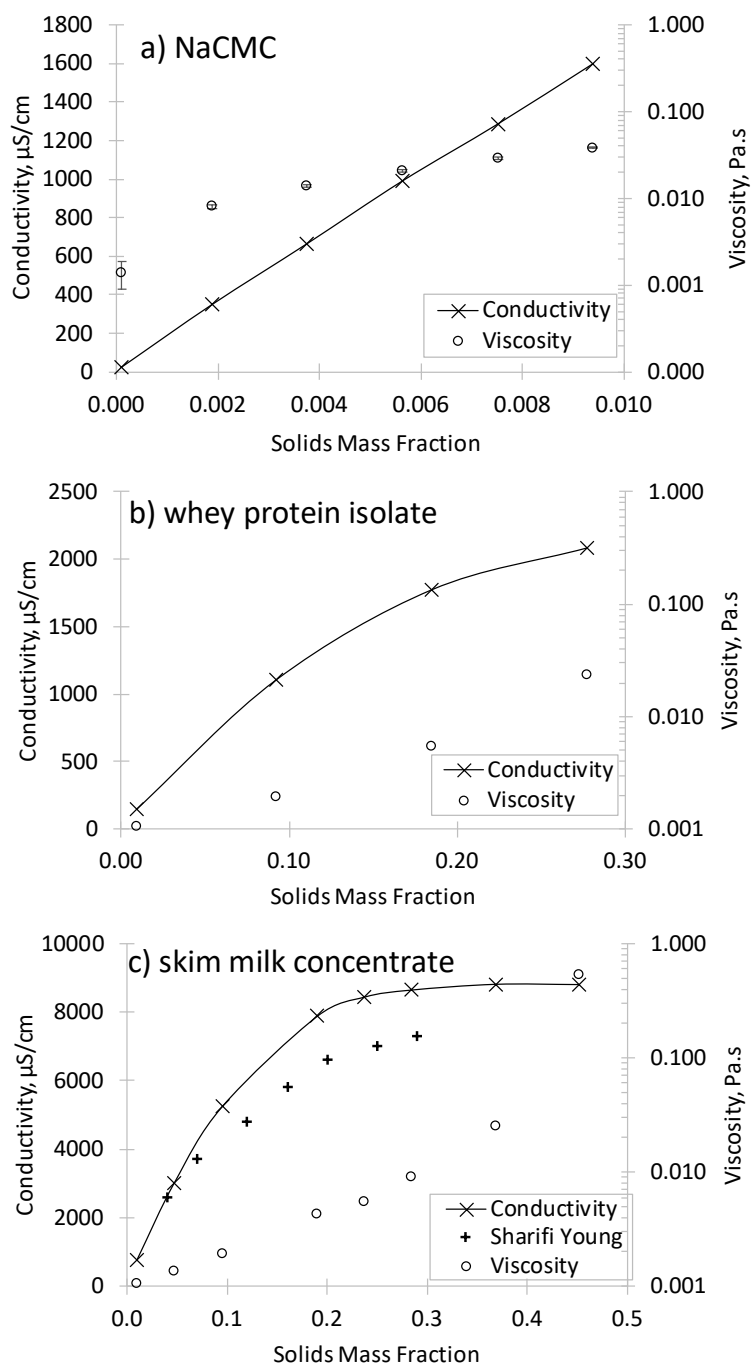
4 Results and Discussion

The relationship between conductivity and viscosity with concentration for four different liquids is shown in Figure 1. In each case the viscosity increases significantly but its effect on conductivity is variable. The conductivity of the NaCMC solution (Figure 1a) is linearly related to concentration and is only slightly affected by the increasing viscosity. The conductivity increases due to the sodium counter-ions and also the residual NaCl normally present in NaCMC. The long NaCMC molecules form a tangled network which increases the bulk viscosity, but within which ions can freely move. The molecular mass of the NaCMC was unknown but it typically about 10^5 g/mol.

At the other extreme is sucrose (Figure 1d) (and other sugars) which show a marked decline in conductivity as the viscosity rises. This has been well studied as stated in the Introduction. The modified Walden equation was fitted giving a value of $\alpha = 0.645$ and a dry basis salt content in sucrose of 39 ppm as NaCl equivalent. The value of α fits well within the range 0.64 ± 0.02 given Miller et al. (2000) for disaccharides. The error bars in Figure 1d were determined from the largest uncertainties found when repeating conductivity measurements of NaCMC, but the deviation from a smoothed curve indicates much lower uncertainties. The uncertainties in other graphs are mostly smaller than the symbols and are shown only in Figure 1a.

The WPI solution (Figure 1b) is similar to NaCMC but there is a reduction in the slope of the conductivity curve. The whey proteins in WPI are β -lactoglobulin and α -lactalbumin, with molecular masses of 18400 and 14200 g/mol respectively, and are small enough to interact more with ions thus reducing their mobility. Skim milk concentrate (Figure 1c) contains about 50% lactose and 40% protein on a dry basis so the resulting conductivity shows behaviour intermediate between the WPI solution and sucrose solution. The near-constant conductivity above 25% solids is most likely caused by the coincidental opposite effects of increased ion

concentration and increased viscosity. The conductivities of skim milk solutions were very similar to those obtained by Sharifi and Young (2012).



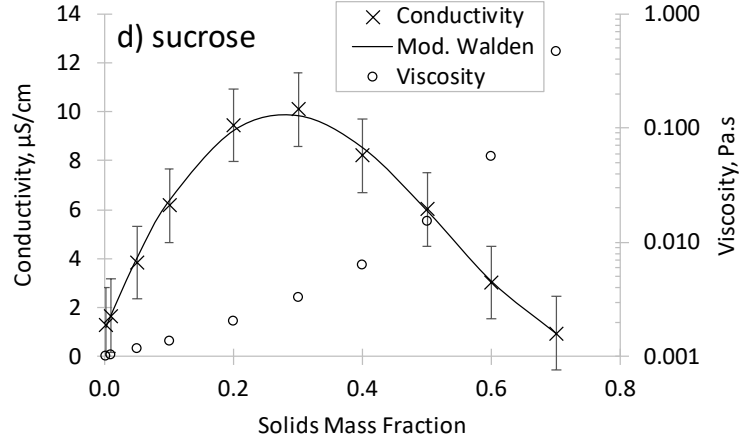


Figure 1. Electrical conductivity and viscosity of four different solutions: NaCMC, whey protein isolate, skim milk powder and sucrose. Conductivities for skim milk solutions from Sharifi and Young (2012) are included in c). The best fit using the modified Walden equation is shown for sucrose in d). Note the logarithmic scale for viscosity.

If the modified Walden Equation (4) is applied to NaCMC, a best fit is obtained using $\alpha = 0.03$, which effectively shows the insignificant contribution of bulk viscosity. It was concluded that Equation (4) is not suitable for NaCMC. Likewise it did not work for WPI and milk solutions.

As an alternative approach the “diffusion viscosity” was calculated using Equation (10). First, using NaCMC + KCl as an example, the equivalent concentration of NaCl in NaCMC was fitted. The conductivity of the equivalent solution of NaCl, as plotted in Figure 2, was fitted using quadratic equations to have the same slope of conductivity as the NaCMC at zero concentrations (where there was negligible viscosity effect). This yielded an equivalent concentration of NaCl of 10.9% dry basis in NaCMC powder. The solution of 0.6% NaCMC + 0.01% KCl was then considered to be equivalent to a solution of $0.006 \times 0.109 = 0.000672$ mass fraction of NaCl + 0.0001 mass fraction of KCl. The conductivity of a mixture of NaCl and KCl at these concentrations was calculated from Equations (6) to (8) to be $1189 \mu\text{S cm}^{-1}$ and the product of the conductivity and viscosity, $f(C)$, of a solution of NaCl + KCl only was $1.192 \mu\text{S Pa s cm}^{-1}$. The measured conductivity was $1151 \mu\text{S cm}^{-1}$ at 20°C . Hence using Equation (10) the diffusion viscosity was calculated to be $1.192/1151 = 0.00104 \text{ Pa.s}$ (only slightly higher than water).

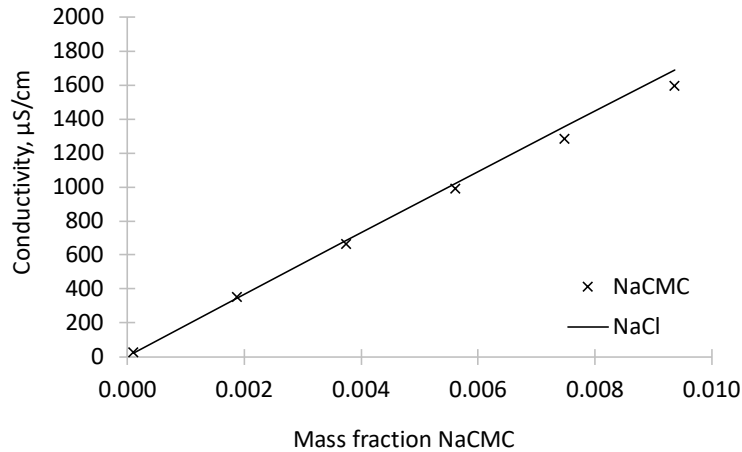


Figure 2. Conductivity of NaCMC solution and of an equivalent solution of NaCl fitted to have the same slope at zero concentration.

The diffusion viscosity was calculated and shown in Figure 3. The first points corresponded to a low conductivity with corresponding large experimental error. The viscosity “experienced” by the conducting ions is shown to be only slightly greater than that of water. Figure 3 shows two different sets of results obtained two months apart, indicating low experimental uncertainty, except at very low concentration where the solution conductivity had greater measurement error.

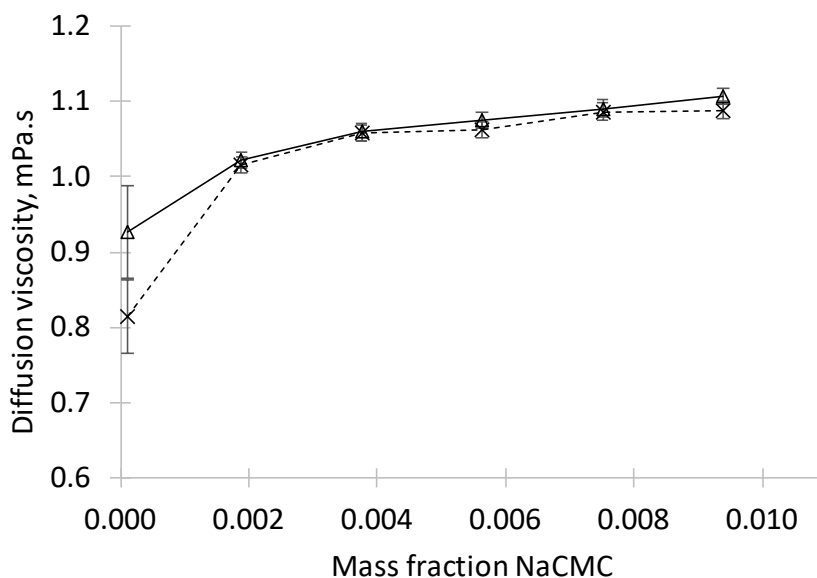


Figure 3. The calculated diffusion viscosity of NaCMC solutions from two different runs. Error bars represent the estimated uncertainty.

Figure 4 shows the result of similar calculations for other mixtures. For comparison, the results for sucrose are included in each graph. The graphs show good consistency between similar solutions, but quite different results for different solutions. The diffusion of ions through NaCMC solutions, with and without salts, seems only slightly affected by mass fraction even though, as seen in Figure 1a, the bulk viscosity increases significantly. Here the mass fractions are very low because higher concentrations are difficult to achieve. The diffusion viscosities for WPI and skim milk are higher than for sucrose at similar concentrations, showing greater resistance to ion flow. This is contrary to expectations as WPI and milk solutions have a lower bulk viscosity than sucrose at the same concentration. It is likely that there is significant interaction between ions and the protein which will have a negative charge in water. The different sugars show similar behaviour to each other.

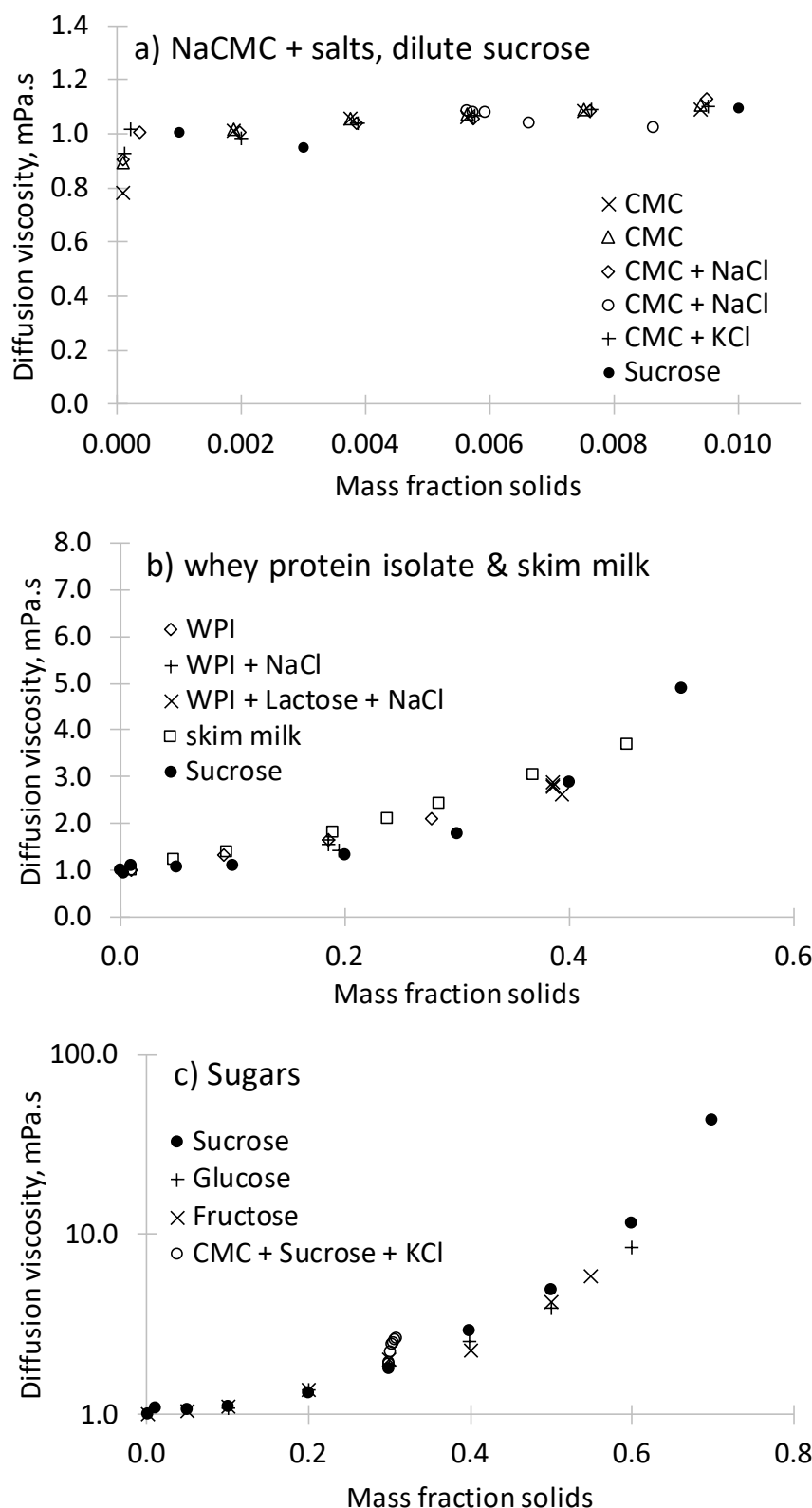


Figure 4. Calculated diffusion viscosity for different solutions. For comparison, results for sucrose are shown on all graphs. Note the log scale for sugars.

The ratio of the diffusion viscosity to the bulk viscosity was calculated and is shown in Figure 5. These graphs clearly show that the bulk viscosity is not a good measure of the viscosity experienced by conducting ions as the ratio is significantly less than 1.0. The indicative error bars of two series show the influence of poor accuracy at low bulk viscosities because of the

accuracy limits of the Haake viscometer at these low viscosities. The relationships for sugars, WPI and skim milk between the ratio and solids content are surprisingly linear with concentration, but given that the relationship involves a number of non-linear equations, this is more likely to be coincidental than fundamental.

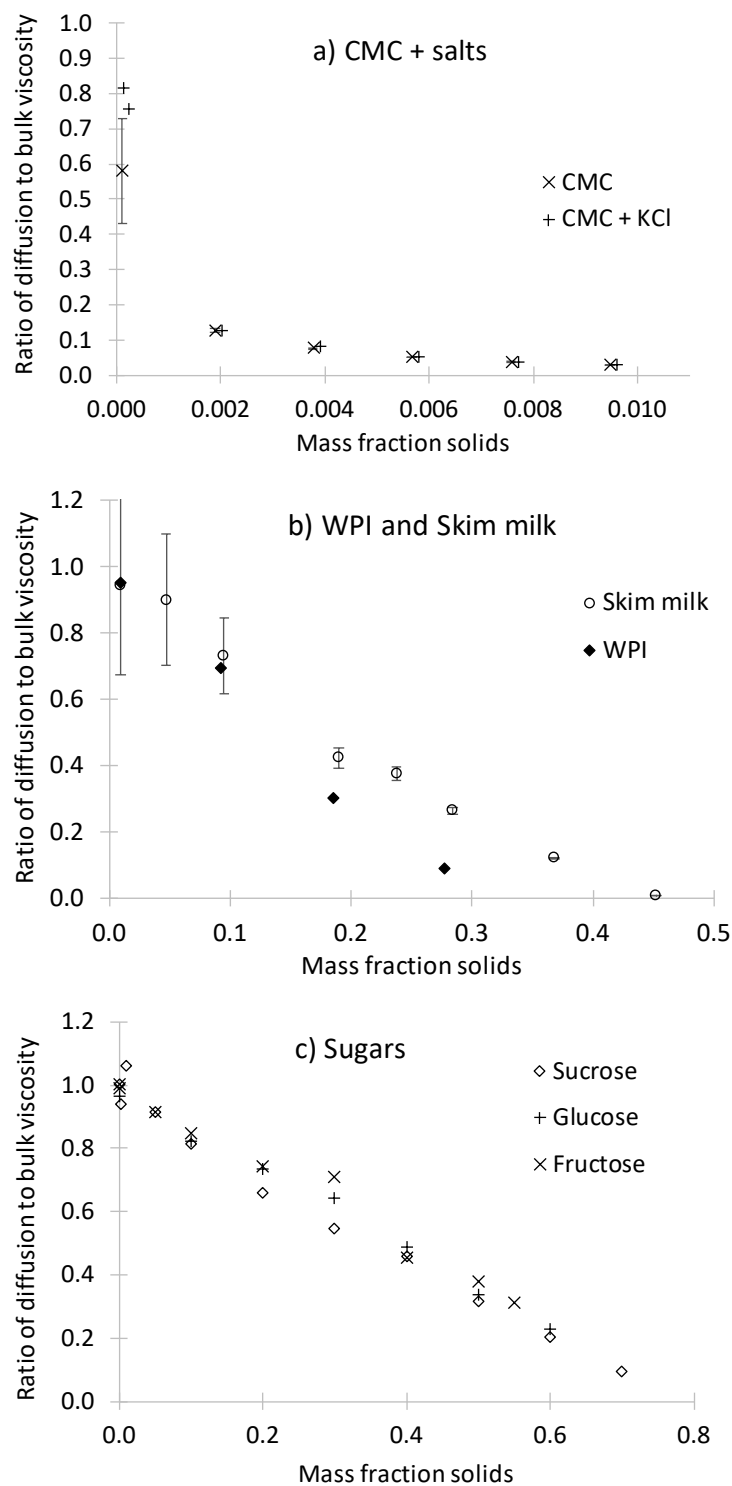


Figure 5. The ratio of calculated diffusion viscosity to bulk viscosity for various solutions. Error bars on two of the series indicate calculated uncertainties.

The results presented so far do not lead to a general prediction equation for electrical conductivity. The modified Walden's rule can be applied to sugar solutions, but the effect of bulk viscosity can probably be ignored in solutions of long chain polymers such as NaCMC. The conductivities of other solutions containing large molecules could not be predicted *a priori*.

For a specific type of liquid it is recommended that the conductivity be related to the diffusion viscosity so that empirical relationships for prediction of conductivity can be determined. However, for a better understanding of ion transport, interactions at a molecular scale could be investigated. For charged molecules like NaCMC and other polyelectrolytes, better predictions for a specific liquid could be obtained by considering that state of the charge and counter-ions, perhaps over a range of pH values. In contrast sugars have minimal interactions with ions and hence a model based on friction between species and tortuous ion diffusion (Krishna and Wesselingh, 1997) might be effective at high concentrations.

When the solids content was low (<10%), the diffusion viscosity was not much greater than that of water, even though the bulk viscosity could be high. Hence the conductivity was mostly dependent on the ion concentration in solution.

5 Conclusions

The application of the modified Walden equation to liquid foods other than sugars was unsuccessful. Experimental results showed that, in general, electrical conductivity was not well correlated with bulk viscosity. Conducting ions were able to pass through viscous NaCMC solutions with little extra resistance. It seems likely that electrical conductivities will be unaffected by the bulk viscosity when the viscosity is high because of entanglement of long polymer chains.

In liquids such as concentrated sugar solutions and honey, in which the viscosity is dominated by small molecules, the electrical conductivity is strongly affected by viscosity. However as already found by many researchers, conductivity of many viscous solutions is not directly related to the inverse of the bulk viscosity.

Instead it was found that a diffusion viscosity could be determined from conductivity data. This showed the resistance to ion movement and gave greater insight into the effects of composition on conductivity.

In milk and whey protein solutions there is some evidence of interaction between ions and the charged proteins which slows the movement of ions and this results in a higher than expected diffusion viscosity. A more complete model including the ion/protein interaction could account for the observed change. Experiments in which protein charge is changed by altering the pH should allow this effect to be quantified.

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